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16. ABSTRACT

The American Meteorological Society, under sponsorship from the U.S. Environmental Protection Agency, the Atmospheric Environment Service, Canada and the Ontario Ministry of the Environment, Canada, convened a workshop in September 1984 to address one aspect of the long range transport modeling which has become a topic of considerable concern, i.e., the quantification of uncertainty in long range transport model predictions.

The workshop was of 3.5 days duration and had participation from 38 invited national and international experts. A series of draft chapters provided a focus for discussion at the meeting and raised the pertinent scientific issues to be addressed by the working groups and to aid in the preparation of findings and recommendations. This summary article is a product of the efforts of all the workshop participations.

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Quantifying Uncertainty in Long-Range-Transport Models:

A Summary of the AMS Workshop on Sources and Evaluation of Uncertainty in Long-Range-Transport Models, Woods Hole, Massachusetts, 18-21 September 1984

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Workshop Chairman

1. Introduction

It has long been recognized that pollutant emissions have the potential of being transported long distances. But it was also generally accepted that the consequences of air-quality impacts due to transported pollutants from distant sources were negligible. To some extent this was justifiable given the diminished concentrations resulting from the dilution and cleansing effects air pollutants undergo during extended transport periods. One practice for the meeting of air-quality standards in the United States, prior to New Source Performance Standards legislated in the Clean Air Act Amendments of 1977, had been to take advantage of dilution effects through the use of tall stacks and the placement of major emission sources in sparsely populated regions with low emission density.

With the advent of increased sophistication in monitoring air pollutants and our continued evolving knowledge of the chemical and physical dynamics of the atmosphere, it has become quite apparent that pollutants can be transported over long distances and have a significant impact on ambient air quality and other environmental media. Documented evidence on the long-distance transport of photochemical oxidants, particulate sulfates, and their respective precursors, as well as the pollutant-related effects of visibility degradation and acidic deposition is now available. As the complexity of the long-range-transport pollutant system became apparent, atmospheric transport and transformation models were developed to help provide an integrated conceptualization of the operative processes.

In addition the models were also used to provide guidance on the relationships between source emissions and pollutant receptors, that is, to provide quantitative estimates of the expected change in air quality or deposition as a result of a specified change in emissions.

The science of air-pollutant simulation modeling plays an important role in elucidating our understanding of the complex chemical and physical interactions of anthropogenic emissions in the atmosphere as well as providing significant guidance as to the most effective strategies to mitigate air-pollutant-related environmental problems. It is therefore important that the scientific community monitors the progress and continues to stimulate this research field to assure that the best science and methodologies are brought to bear on the problem.

To this end, the American Meteorological Society (AMS), under sponsorship from the U.S. Environmental Protection Agency, the Atmospheric Environmental Service, Canada and the Ontario Ministry of the Environment, Canada, convened a workshop in September 1984 to address one aspect of long-range-transport modeling that has been a topic of considerable concern, the quantification of uncertainty in long-range-transport (LRT) model predictions.

2. Workshop

The workshop was three and one-half days in duration and had participation from 38[2] invited national and international experts. This summary report is a product of the efforts of all the workshop participants. The workshop had the following objectives: 1) to examine current understanding of the physical processes responsible for long-range transport and depo-

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[2] The participants were as follows: Dr. Len Barrie,* Atmospheric Environment Service; Dr. Bruce Beck, Imperial College of Science & Technology; Dr. Chandrakant Bhumralkar, SRI International; Dr. Gregory Carmichael, University of Iowa; Dr. Julius Chang,** National Center for Atmospheric Research; Dr. Han-Ru Cho, University of Toronto; Dr. Al Christie,* Atmospheric Environment Service; Mr. Terry L. Clark, NOAA/MD ASRL; Dr. Kenneth L. Demerjian,* Harvard University; Mr. Roland Draxler, NOAA/ARL; Dr. Anton Eliassen, Norwegian Meteorological Institute; Dr. James A. Fay, Massachusetts Institute of Technology; Dr. Bernard Fisher, CERL/England; Dr. Douglas Fox,* USDA Forest Service; Dr. Robert Friedman, Office of Technology Assessment; Dr. Jerome Hales, Battelle PNL; Mr. Bruce Hicks, NOAA/ATDL; Dr. Daniel Jacob, California Institute of Technology; Dr. Oystein Hov, Norwegian Institute for Air Research/NILU; Dr. Frederick Lurman, Environmental Research & Technology, Inc.; Dr. Daniel McNaughton, TRC Environmental Consultants, Inc.; Dr. Gregory J. McRae, Carnegie-Mellon University; Dr. P. K. Misra,* Ontario Ministry of Environment; Mr. Marvin Olson, Atmospheric Environment Service; Dr. Lars Prahm, Danish Meteorological Institute; Dr. Philip Roth,* Consultant; Dr. Ian Rutherford, Atmospheric Environment Service; Dr. Perry Samson,** University of Michigan; Mr. Frank Schiermeier,* NOAA/MD ASRL; Dr. Jack D. Shannon, Argonne National Laboratories; Dr. George Slinn, Battelle PNL; Dr. Akula Venkatram,* Environmental Research & Technology, Inc.; Dr. Eva Voldner, Atmospheric Environment Service; Dr. Thomas Warner, Pennsylvania State University; Dr. Boris Weisman, MEP Company; and Dr. Fred White,* American Meteorological Society.

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sition of atmospheric pollutants, with emphasis given to the uncertainty in our understanding of these processes; 2) to examine the parameterization of these processes in available long-range-transport models and to estimate the effects of parameterizations on model uncertainty; 3) to examine the temporal and spatial scales of applicability of the available long-range-transport models; 4) to examine the data base available for use in long-range-transport modeling and, to the extent possible, estimate the limits of predictability associated with the use of this data base; 5) to recommend methods to test long-range-transport-model results with corresponding observations with a view to quantifying model uncertainty; and 6) to produce a report summarizing the workshop findings.

A series of invited draft chapters were prepared and distributed to discussion leaders several weeks prior to the meeting. The draft chapters provided a focus for discussion at the meeting and were intended to raise the pertinent scientific issues to be addressed by the working groups and to aid in the preparation of findings and recommendations. The workshop-report refinement process occurred via the plenary discussions and working-group sessions held throughout the three and a half days.

3. Draft chapters

The titles and authors of the five draft chapters prepared for the workshop were as follows: "A Policy Perspective on Long Range Transport Models"—Robert M. Friedman; "Sources of Error and Uncertainty in Eulerian Long Range Transport Models"—Gregory R. Carmichael; "Sources of Uncertainty in Lagrangian Long Range Transport Model Calculation"—Anton Eliassen; "Stochastic Long Range Transport Models"—Akula Venkatram; "Methods for the Sensitivity and Uncertainty Analysis of Long Range Transport Models"—Gregory J. McRae.

An AMS publication containing the complete documentation of the workshop including the revised chapters, recent developments in model evaluation and sensitivity analysis, and the full reports of the Eulerian and Lagrangian working groups is in preparation.

4. Findings and recommendations of the Eulerian and Lagrangian working groups

The findings and recommendations of the Eulerian and Lagrangian working groups showed similarity and generic focus in several areas. One of these areas in particular was ranked highest in priority by the two groups. The consensus of the two working groups indicates that the number one priority with regard to the elucidation and quantification of uncertainty in LRT models is the development of improved data bases for model testing and performance evaluation. For the sake of clarity and brevity these results have been combined and presented first in this section.

5. Data requirements for model-performance evaluation

It was the general consensus of the United States/Canada (U.S. Environmental Protection Agency, 1983) modeling subgroup that part of the differences between the eight model results was the result of different processing of the meteorological input data and the quality of that data. Future improvements in the ability of LRT models to reproduce observed concentration and deposition fields will depend in a large part upon more detailed and quality-assured data bases. Specifically, the data bases that will need to be improved are: 1) emissions; 2) meteorological; and 3) chemical.

a. Emissions

The ability of LRT models to predict daily and/or event concentration and deposition values is linked to knowledge of the corresponding fluctuations in emissions rate for SO_2 and other pollutants. The daily emission values for the largest sources in eastern North America should be collected and archived along with an analysis of monthly-mean diurnal and weekday/weekend emission patterns for smaller sources. Nonlinear chemical-transformation processes also require information about the emissions of nitrogen oxides, reactive hydrocarbons, and ammonia. This emission information should be obtained on an annual basis at first with analysis of monthly and weekday/weekend variations.

b. Meteorological measurements

The primary set of measurements should be obtained on a nested grid with varying resolution ranging on the course scale from 200 km and 6 h to that on the fine scale of 40 km and 1 h. They include:

- ground-level precipitation;
- cloud type and spatial characteristics;
- vertical profiles of temperature, humidity, and wind;
- vertical profiles of cloud-water and hydrometeor concentrations.

In addition, detailed intensive boundary-layer studies should be performed in concert with the basic research-monitoring program to acquire highly resolved concentration and turbulence data in conjunction with relevant biological properties effecting the dry deposition of atmospheric species.

c. Chemical measurements

Ambient measurements of a wide variety of chemical species are required to run and evaluate regional air-quality and deposition models that incorporate sophisticated descriptions of the chemical-transformation processes occurring in the atmosphere. In addition, information on the emission of a select number of chemical species is also required. A list of the chemical species, measurement and monitoring characteristics, and rationale for collection is presented in the full report.

The averaging time in regional models is of the order of 1–3 h, which is commensurate with the models' spatial resolution of 20–100 km. As with the meteorological data, nested monitoring networks of varying resolution will provide the

necessary data to understand and develop improved techniques, if warranted, to treat subgrid-scale chemical and physical processes. In addition, enhanced spatial resolution of the monitoring data provides the opportunity to determine the spatial representativeness of point measurements, an issue of importance not only to the performance evaluation of models, but to the empirical estimation of depositional loading itself.

6. Eulerian LRT model expectations

Eulerian long-range-transport (ELRT) models are designed to study the relationship between source emissions and receptor air quality and deposition over large areas. At present there are several large-scale development activities directed at constructing models of the physical and chemical processes occurring in the atmosphere. Given the availability of suitable input data it is expected that these models will be able to predict:

1. Gas-phase concentrations of: NO_x, SO_x, O_3, NO_2, H_2O_2, PAN, HONO, NH_3, HNO_3, reactive hydrocarbons and CO;
2. aqueous-phase concentrations of: H^+, $SO_4^=$, NO_3^-, NH_4^+, organic acids, and chloride;
3. aerosols, with respect to simple equilibrium formulations of the sulfate-nitrate-ammonia system. (A clear direction for future development activities would be to add the capability to handle particle-size distributions.)
4. All the chemical species at a horizontal resolution of approximately 100 km × 100 km. (Predicted fields, over large dimensions for use in analysis and interpretation, can be calculated by simply accumulating the gridded information.)
5. Species concentrations with a time resolution as small as the numerical integration constraints; that is, time steps of order of 10 minutes. (Longer-time-averaged concentrations of predictands for comparisons against observations can be carried out by temporally averaging the model outputs.)

Finally, given the gas and aqueous species, described above, it will be possible to calculate both the dry and wet deposition patterns over regional scales as well as at target receptor locations with these models.

7. Criteria and procedures for quantifying errors in Eulerian LRT models

It is the opinion of the working group that the appropriate criteria and procedures for quantifying errors in ELRT models should be based on two considerations: 1) the need to demonstrate effectively the impact of alternative control strategies, and 2) the need to quantify indices of model performance over the total domain or over subdomains representative of different meteorological processes. The former could be semiquantitative (relying on relative signal changes) until experience with the models permits determination of a more objective numerical criterion. The latter would make use of measures of the models' precision in simulating time and space variations in model output parameters to guide in systematic improvement in process representation.

The uncertainty in model outputs consists of input uncertainty and model-formulation uncertainty. The formulation uncertainty will include a component relating to the representation of the physical-chemical processes and partly attributable to the mathematical integrations. Insight into the models' response to arbitrarily varying inputs for a particular formulation can be studied effectively by methods currently available (Rabitz *et al.* 1983; Tilden *et al.*, 1981; Tilden and Seinfeld, 1982). Complete evaluation of the models demands systematic comparison with quality-controlled observational data over as extensive a model domain as possible. It should be recognized that the effect of subgrid-scale processes, turbulence representation in transport processes, and problems of specification of input fields will result in a limit of predictability analagous to that for meteorological forecast models.

Model evaluation initially should emphasize the time and space scales simulated by the LRT models by studies of: 1) standard statistical measures of predictands previously mentioned over the total domain and over stratified data subsets; 2) time series of model-observation residuals; 3) and pattern-recognition techniques.

There is a further need to establish the bias and uncertainty associated with averaging or integrating model outputs as required as input to evolving environmental-effects-type models.

Currently available data sets such as OSCAR (Easter *et al.*, 1984) and NEROS (Schere and Fabrick, 1985; Vaughan, 1985) need to be studied in depth, in conjunction with improved emission inventories, to give preliminary insight into evaluation methods and to refine design factors of a more comprehensive evaluation data base such as length of record, numbers of replications, data-stratification methods, and model-parameter-specification methods. Finally there is a need to examine in detail how the model-performance criteria should be formulated.

8. Eulerian model outputs for long time averages

Current-generation ELRT models, by nature of their comprehensiveness and complexity, are the most applicable and economically efficient in studying episodic acid-deposition events, i.e., events lasting a few days to a week. In principle, given the proper continuous input data, these models can estimate seasonal or even annual averages of acid deposition directly. To do so would be very costly computationally. Several procedures have been suggested to circumvent this difficulty.

First, it has been observed that many of the synoptic events in given regions and over selected time periods, for example the northeastern United States and southeastern Canada on an annual basis, can be grouped into general meteorological categories. It is proposed that ELRT models can be used to study the deposition associated with each category of synoptic event and then use the cumulative simulation data to estimate seasonal and annual averages. In establishing the frequency

of occurrences of each category of deposition event in a season and the amount and distribution of deposition, one can compute a statistical ensemble average for that season. Unfortunately, this procedure has not seen prior application in meteorology or in air-pollution research on a regional scale. A feasibility study should be carried out with one or more existing data bases. Sulfate wet deposition in the eastern United States is a possible choice for testing the technique.

A different approach toward obtaining seasonal and annual averages would be the use of full-scale ELRT models to develop simpler models that are more computationally efficient but less comprehensive in the inclusion of detailed physical and chemical processes. These simpler models, or "engineering models," can be either Eulerian or Lagrangian in structure. Their essential feature must be that the accuracy of their representation of the physical and chemical details can be verified by selective use of the comprehensive ELRT models. These computationally efficient engineering models then will be used to study seasonal and annual depositions on a regional scale. It is emphasized that simulation results from these engineering models must be checked frequently to avoid significant deviations from the results of the comprehensive models and from relevant observational data.

9. Uncertainties in Eulerian model predictions

A large contribution to the uncertainty in ELRT model predictions is due to the spatial and temporal limitations of the available input data. It is generally felt that model uncertainty can be reduced by increasing the spatial and temporal resolution of the input data (e.g., rawinsonde data, land-use information, and emissions). However, since the cost of obtaining higher-density and more-frequent inputs may be large, it is important to estimate how the uncertainty will decrease as the input resolution is increased. One approach to quantifying the improvement is to use existing models to study their sensitivity to input data. For example, the NCAR meteorological model is being used to investigate the sensitivity of predicted trajectories to changes in the spatial and temporal resolution of the wind field.

It is recommended that direct-simulation studies, using existing models, be initiated to quantify the sensitivity of model predictions to increases in the spatial and temporal resolution of the input data, bearing in mind that, in the absence of prior model testing against reliable data, the derived output sensitivity may reflect partly model artifact.

10. The focus of future research to reduce uncertainties in Eulerian models

The largest sources of uncertainty in current LRT Eulerian models are due to lack of data bases to initiate, run, and evaluate the model and its modules and to incomplete formulation of processes in models. Specifications for an observational program using existing instrumentation have been outlined in a previous section of this paper. In order to improve the quality of such observations it is necessary that considerable research and development efforts be directed toward improving the sensitivity, time resolution, and robustness of air-concentration measurement techniques especially for sulfur-aerosols, SO_2, NO_x, NH_3, PAN, H_2O_2, and reactive hydrocarbons.

Better formulations and evaluation data bases are needed for subgrid-scale representation of the following processes:

1) transport/diffusion, removal, and chemical-conversion processes associated with cloud fields including the very neglected, important process of fog-water deposition;
2) dry-deposition processes, in particular those related to complex terrain and heterogeneous, biologically controlled surfaces.
3) the transport/diffusion, removal, and chemical conversion of pollution released by point or area sources smaller than the grid resolution of the model.

Another model formulation that is unnecessarily incomplete is that for aerosol species, which are not distributed by particle size in current-generation regional models. Since the processes of wet and dry removal and that of chemical transformation are particle-size dependent, the incorporation of an aerosol-growth module in the models is an important improvement for consideration.

There is insufficient information on the mechanism of air-surface exchange of many gases in the model (e.g., hydrocarbons). It is recommended that further experimental studies as well as exercises that generalize experimental results be undertaken.

There are several research needs related to gas-phase chemistry uncertainties: 1) more smog-chamber studies on time scales applicable to LRT schemes; 2) investigation of nighttime chemistry of nitrogen compounds; 3) kinetic data for temperatures of 0–35°C rather than the more typically studied temperature of 25°C; and 4) continued study of the role of natural hydrocarbons in the photochemical process.

It is recommended that methods for obtaining seasonal- or annual-mean concentration fields, deposition fields, and source-receptor transfer matrices be investigated. Two possibilities are: 1) to use the comprehensive Eulerian model to obtain simple, accurate parameterizations of processes that can be used in the faster Lagrangian or "engineering-type" Eulerian models to generate long-term average outputs; and 2) to study the feasibility of stratifying meteorological synoptic classes, from which analysis of a fraction of representative classes can be used to reconstruct seasonal or annual outputs (means, totals, distribution functions, etc.).

Another important task related to transport mechanisms is to improve four-dimensional data-assimilation procedures. A large part of the uncertainty in the transport/diffusion calculation in an LRT model is due to the lack of meteorological information on the mesoscale. Fundamentally, there is a lack of mesoscale data, but to a considerable extent the gaps can be filled through the use of a mesoscale meteorological model incorporating good boundary-layer treatment. The assimilation and initialization must be done in such a way as to incorporate the synoptic-scale information present in the data and at the same time preserve the smaller-scale structure produced by the model.

11. Lagrangian LRT model applications and expectations

Lagrangian long-range transport (LLRT) models have been used widely both to diagnose source-receptor relationships for atmospheric sulfur oxide concentrations and sulfur deposition, and to estimate the potential changes in sulfur concentration and deposition values following changes in the emissions distribution of sulfur dioxide. Similarly, some LLRT models have also been used to calculate the concentrations and deposition of nitrogen compounds. In general, the LLRT models are popular because: 1) they are simpler in concept than ELRT models, and generally more inexpensive to operate, and 2) they are presently operational. Several reviews of LLRT models have been published (Bass, 1980; Eliassen, 1980; Johnson, 1981; Fisher, 1983). The reader is referred to these articles for summaries of the methodologies used in LLRT models.

Lagrangian long-range transport models have demonstrated an ability to reproduce long-term (annual) SO_2 and particulate $SO_4^=$ concentrations that are in reasonable agreement with observations. The least complicated of the LLRT models, the so-called semi-empirical models, have been shown to reproduce annual $SO_4^=$ concentrations to within a factor of 2 both in the United States (Venkatram *et al.*, 1982) and in Europe (Fisher and Clark, 1984). Likewise the more deterministic LLRT models have also been successful in reproducing observed annual-average $SO_4^=$ concentrations. Eliassen and Saltbones (1983) in Europe and Shannon (1981) in North America have shown skill in predicting annual-average particulate $SO_4^=$ concentrations.

There is also a statistically significant, albeit less satisfactory, agreement between calculated and observed weighted-mean annual-average concentrations of $SO_4^=$ in precipitation. The reduction in accuracy in predicting the sulfur wet deposition must be viewed relative to the accuracy of sulfur-wet-deposition measurements. In order to assess the degree of uncertainty in model predictions of concentrations and deposition, Fox (1984) has concluded that it is necessary to study the variance of the observations in relation to the variance of the residuals (differences between observation and prediction). Voldner and Shannon (1983) predicted the annual wet deposition of sulfur in eastern Canada within the uncertainty of the observations. Eliassen and Saltbones (1983) predicted the two-year average sulfate concentration in precipitation at 33 sites with a correlation coefficient of 0.70 as compared to a correlation coefficient of 0.85 for ambient sulfate. The reduction in accuracy in predicting sulfur wet deposition is believed to be related to the episodicity of precipitation events.

On a daily basis LLRT models can give some useful information on SO_2 and particulate $SO_4^=$ concentration, but their strength is in the reproduction of the distribution of daily concentrations over a year rather than in pair-wise agreement. The comparison of daily estimates of SO_2 and $SO_4^=$ with observations also serves the purpose of identifying meteorological regimes during which model performance is improved and regimes during which it is poor. While pair-wise agreement between predicted and observed concentrations does not in itself guarantee reliable estimates of source-receptor relationships, it does give us more confidence in defining such relationships when the agreement is high. Voldner *et al.* (1981), Bhumralkar *et al.* (1981), and Samson and Small (1984) have demonstrated the ability of the models to reproduce frequency distributions of $SO_4^=$ concentrations. The pair-wise correlation of predicted and observed daily $SO_4^=$ has been reasonably high in some studies (eg., Voldner *et al.*, 1981; Eliassen and Saltbones, 1983); but poor in others (Ruff *et al.*, 1984). In one model evaluation, a Lagrangian long-range-transport model demonstrated more skill in simulating temporal variability than in simulating the fine detail of spatial concentration distributions (McNaughton *et al.*, 1981).

The LLRT models have not demonstrated an ability to reproduce sulfur-wet-deposition fields on a daily or event basis as yet. This is probably due in part to the oversimplification of pollutant scavenging processes in the models, but more basic problems exist that limit our ability to ascertain specific reasons for the poor performance. The episodic behavior of precipitation events, the fact that the two-dimensional trajectories computed for precipitation events may have limited reliability in the vicinity of fronts, and our incomplete knowledge of scavenging processes undoubtedly limit the accuracy of the models' predictions of short-term sulfate deposition.

There are actually few essential differences between Lagrangian and Eulerian long-range-transport models apart from the frame of reference with which the flow is treated. The simplicity of LLRT models forces trade-offs that limit their resolution relative to ELRT models. The specification of nonlinear chemical reactions, for example, is possible with an LLRT model, but the implied horizontal mixing of the LLRT model limits the resolution of impact from specific source regions. Likewise, vertical resolution of concentration fields is more easily obtained through ELRT-modeling techniques. If the Lagrangian models are reformulated to include all major effects of stratification within the air parcel, the advantages associated with their simple and elegant basic formulation are quickly lost and replaced by prohibitive complexity. This implied rapid vertical mixing in LLRT models influences the calculation of mass flux of material to the surface through dry deposition.

The description of horizontal dispersion due to vertical wind-velocity shear over long time scales is intrinsically ignored in LLRT models. Samson (1980) described an uncertainty field about the mean flow resulting from the average effect of velocity shear on trajectory location, but rapid horizontal mixing of the air within the domain of this field is nonetheless presumed. Heffter (1983) has developed a branching trajectory model that splits trajectories in response to nocturnal deformation of the daytime transport layer.

Eliassen *et al.* (1982) and Rodhe *et al.* (1981) have demonstrated that nonlinear atmospheric chemical processes can be included in receptor-oriented LLRT models, but this assumes that the rate of horizontal mixing of air into the Lagrangian column or puff is known and that the boundary conditions for each species carried is known.

12. Recommendations for improvement in Lagrangian LRT models

There are several possibilities for improvement in LLRT models. In addition to those already noted regarding data im-

provements and advances in understanding of relevant processes, the following improvements in LLRT modeling in order of decreasing priority should involve: 1) improvements in the analysis and interpretation of available input data; 2) more-realistic parameterization of scavenging processes; 3) more-realistic representation of chemical-transformation processes; 4) more detail on nonhomogeneous nature of dry deposition; 5) inclusion of cloud venting. These will be discussed individually below.

a. Improvements in the analysis and interpretation of available input data

The ability of LLRT models to reproduce sulfur-wet-deposition values is primarily a function of the model input to reproduce the amount and type of precipitation occurring at the receptor and along the trajectory. Keeler *et al.* (1984) demonstrated that the ability of an LLRT model to reproduce sulfur wet deposition was limited by the accuracy of the precipitation input data. The methods used to grid precipitation data, particularly in data-sparse regions, should be evaluated for representativeness. McNaughton (1984a) has demonstrated the problems involved in preparing hourly grids of precipitation even in relatively data-rich areas and how misspecification of the precipitation inputs can account for errors in wet-deposition estimates (McNaughton, 1984c).

Information on cloud type, cloud thickness, cloud-base height, and percent coverage along the trajectory will be useful to the improved parameterization of several processes in long-range transport modeling including heat-flux estimation (for aerodynamic resistance calculations), aqueous-phase chemical transformation, and wet scavenging. Although cloud data bases do exist with reasonable spatial and temporal resolution (e.g., Fey, 1978), enormous data processing will be required to use this information. A second choice for LLRT models will be to use meteorologically stratified climatologies of cloud occurrence.

The wind-field information currently used in most LLRT models is derived from twice-daily upper-air observations made by synoptic networks. It is recognized that an improvement may be possible through the use of numerical weather prediction (NWP) model-wind-field outputs. Kuo *et al.* (1984) have illustrated an increase in trajectory accuracy using NWP-analyzed winds versus normal, twice-daily observations. The NWP model would, in effect, be used as a sophisticated temporal- and spatial-interpolation methodology for winds and thermodynamic properties which would be mass-consistent and dynamically correct, more reliable in data-sparse areas, and, potentially, more descriptive of phenomena such as the nocturnal jet.

It is recommended to improve LLRT model predictions of dry-deposition processes by including more detail on land-use patterns. More detail on land-use patterns in Canada, especially in remote regions (Sirois and Voldner, 1984), coupled with estimates of the atmospheric stability and surface resistances, may yield better estimates of the rate of dry deposition in those areas.

b. More-realistic parameterization of scavenging processes

Better realization of the scavenging rates for SO_2 and $SO_4^=$ in LLRT models may be gained through better parameterization of the ensemble cloud physics and chemistry associated with entrainment, nucleation, and cloud-droplet solubility. Such parameterizations may be based on a priori assumptions of the scavenging process or may be linked to estimates of the environment of other pollutants in the interstitial air of the clouds. Barrie (1981), for example, has suggested that the SO_2 washout ratio could be described as a function of rainwater pH and temperature. It may also be possible to describe the scavenging of SO_2 as a function of cloud-water hydrogen peroxide (H_2O_2) whose concentration could itself be parameterized as a function of cloud type, latitude, season, and/or other atmospheric parameters.

It is important that the parameterization of scavenging, when based on extrapolation of theory, be reviewed for consistency with the original theory. McNaughton (1984b) has illustrated common overextension of the original theory of Scott (1978) in some current Lagrangian long-range-transport models.

c. More-realistic representation of chemical transformation processes

More-realistic representation of both gas-phase and aqueous-phase chemical-transformation processes will yield more confidence in the use of LLRT models for predicting concentration and deposition changes resulting from changing emissions patterns. The chemical mechanisms that will be used in LLRT models will necessarily be highly parameterized, reflecting the essence of chemical interrelationships gleaned from the development of more-sophisticated chemical modules for ELRT models.

Aqueous-phase transformation will require more information on cloud type, depth, base height, and percent cover than is presently used in LLRT models. There will also need to be a coupling of the gas-phase and aqueous-phase processes through some explicit assumption concerning the cloud-entrainment rates. Gas-phase chemistry will also require some knowledge of the cloud field in order to estimate the amount of sunlight available along the course of the trajectories for photochemistry.

d. More detail on nonhomogeneous nature of dry deposition

For transport over long distances, it becomes very important to know the deposition velocities for the various species accurately. Sophisticated parameterization of the process requires considerable detail in atmospheric and surface resistances to uptake of the species. While the atmospheric resistance can be estimated reasonably well, field experiments show that the surface resistance for important gases such as SO_2 and O_3 exhibit a large, surface-specific variation. For many other gases and particles, existing information is insufficient.

Improvement in the description of aerodynamic resistance in Lagrangian modeling is also possible. This improvement may be accomplished through a parameterization of the heat flux using time-of-day and cloud cover as surrogates for solar-intensity measurements. A critical research area for many Lagrangian modelers will be the development of simple deposition parameterizations that are consistent with more-sophisticated treatments.

e. Inclusion of cloud venting

It is estimated that, in certain circumstances, convective activity will be sufficient to redistribute pollutants from the mixed layer to a higher elevation. LLRT models could better represent surface concentrations of pollutants if upwind venting were included. Unfortunately, LLRT models are inadequate for re-entraining vented air into the mixed layer on subsequent days without assuming the vented material to be spatially homogeneous.

13. LLRT model uncertainty and the user

In order to assess the uncertainty in models and enhance the credibility in model predictions, it is essential that model results be compared with network data. For a meaningful intercomparison between predicted and observed quantities it is necessary that the errors and uncertainties in the observations be quantified. In addition, one should carefully consider the statistical measures to be used and how to interpret subsequent results. Every effort should be made to encourage model intercomparison and evaluation activities. One such example, the International Sulfur Deposition Model Evaluation (ISDME), cosponsored by Environment Canada and the U.S. Environmental Protection Agency (Clark *et al.*, 1984), will compare the results of seasonal and annual wet-deposition amounts predicted by several European and North American long-range transport models with deposition amounts estimated from networks in eastern North America.

One of the principal uses of the LLRT is the establishment of source-receptor relationships. Thus, it would also be desirable to evaluate the ability of models to reproduce these relationships. Unfortunately, there is little data for this purpose. Presently, the ability of a model to reproduce source-receptor relationships is circumstantial, based on the ability to reproduce observed concentration patterns. Future field studies are needed 1) to evaluate our ability to reproduce the trajectory of the winds through use of passive, nonreactive tracers and 2) to evaluate the parameterizations of deposition with tracers that emulate the pollutants of interest.

Lagrangian long-range-transport models represent *reasonable* approximations to complex atmospheric processes. The user of model predictions must, however, keep in mind the spatial limitations imposed by the LLRT framework. Attempts to apply LLRT models to near-field deposition or to situations where vertical stratification of pollutants occurs exceed the capability of these models. Further, the application of these models with linear chemical transformation to future emission scenarios must also be performed cautiously until more-complete chemical mechanisms can be incorporated and tested.

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